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Optimization of moving bed biofilm reactors for oil sands process-affected water treatment: The effect of HRT and ammonia concentrations

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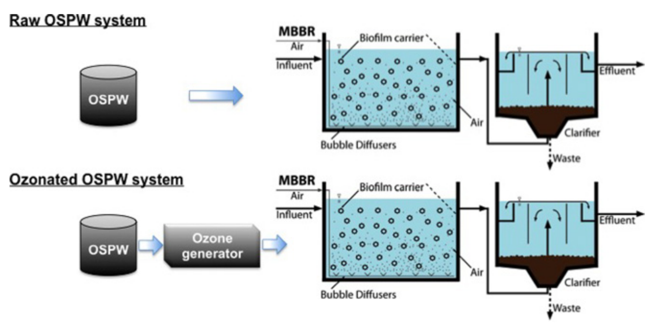
HIGHLIGHTS

- MBBRs were optimized by changing HRT and ammonia concentrations for OSPW treatment.
- High ammonia levels were more beneficial than an extended HRT for OSPW treatment.
- Both AEF concentration and composition were relevant to the toxicity of OSPW.
- An increase in ammonia level favors biodegradation of high molecular weight NAs.

GRAPHICAL ABSTRACT



Schematics of raw and ozonated OSPW MBBRs

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ABSTRACT

Two moving bed biofilm reactors (MBBRs) were optimized to improve the biodegradation of organic compounds in raw and ozonated OSPW by changing the hydraulic retention time (HRT) and the influent ammonia concentrations. During the five stages, the average COD removal reached 50.8 ± 3.4%, 52.8 ± 6.5%, 54.7 ± 4.3%, 56.3 ± 2.2%, and 58.0 ± 2.3% respectively in raw OSPW MBBR, and 54.6 ± 3.8%, 57.2 ± 7.1%, 55.5 ± 5.8%, 58.3 ± 2.2%, and 60.7 ± 2.3% respectively in ozonated OSPW MBBR. Welch's weighted ANOVA tests show that the increase in ammonia levels significantly improved the COD removal in the two systems, while the HRT was an important parameter for COD decrease in the raw OSPW MBBR. Compared to the HRT, the increase in ammonia concentrations were more beneficial for acid extractable fraction (AEF) degradation and the average AEF removal reached 29.80% (raw OSPW MBBR) and 16.50% (ozonated OSPW MBBR) by the end of the optimization (Stage V; HRT = 96 h, 60 mg/L NH_4^+-N). >98% of the NH_4^+-N was removed in the two MBBR systems, showing good nitrification. Microtoxicity tests showed that no significant correlations were found between HRT/ammonia levels and the OSPW toxicity changes toward *V. fischeri*. Spearman's rank correlation analysis was applied for *q*-PCR data, showing that positive correlations between the removal efficiencies of AEF and NSR and NirK gene copies were observed in the raw OSPW MBBR system, while positive correlations between AEF removal efficiency and total bacteria gene, NSR, Nitro, and NirK gene copies were observed in the ozonated OSPW MBBR system.

1. Introduction

Bitumen extraction from the oil sands leads to a large quantity of oil sands process-affected water (OSPW). The acute toxicity of OSPW has

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been a crucial environmental concern in recent years (Lo et al., 2006). OSPW is a slightly alkaline mixture of petroleum hydrocarbons, organic acids, salts, heavy metals, and suspended solids (Huang et al., 2015). Due to a zero-discharge policy established by the Alberta government, large amounts of OSPW accumulate in on-site ponds, where its toxic components (referred to as tailings) kill or inhibit zooplankton, plants, and bacteria (Clemente et al., 2004). Remediation of OSPW is urgently needed to allow OSPW to be recycled or safely discharged to the environment.

Organic naphthenic acids (NAs), a broad group of alkyl-substituted acyclic and cycloaliphatic carboxylic acids (Choi and Liu, 2014; Martin et al., 2010), are the primary toxic constituents of OSPW (Rogers et al., 2002; Scott et al., 2005). Values of the organic acid extractable fraction (AEF) in OSPW are usually used for the evaluation of the quantity of NAs and other compounds containing ketones, carboxylic acids, as well as aldehyde functional groups (Jivraj et al., 1995; Zhang et al., 2016). NAs have an in situ biodegradation half-life of around 13 years (Han et al., 2009). Since the endogenous microorganisms cannot meet the urgent need for OSPW treatment (Shi et al., 2015), an efficient process to biodegrade the toxic components in OSPW is the main focus of many researchers.

Bioreactor technology is an economical, energy-efficient, and environment-friendly approach to wastewater treatment. In bioreactors, extracellular polymeric substances (EPS) secreted by cells protect microorganisms against environmental stress and also act as a nutrient source; within the context of the EPS matrix, microorganisms can act synergistically. OSPW toxicity may affect bioreactor operation and hereby the selection of reactor configuration that encourage the growth of slow growing NAs degraders and facilitate the growth of microbial forms that are resistant toward toxicity should be considered when designing biological treatment of OSPW (Shi et al., 2013). Engineered bioreactors, in which suspended or/and attached growth biomass are applied, have shown high NAs removal efficiency in OSPW (Huang et al., 2015; Shi et al., 2015; Xue et al., 2015). Huang et al. (2015) removed 43.1% of the NAs concentration in raw OSPW with hydraulic retention time (HRT) of 48 days using integrated fixed-film activated sludge (IFAS) reactors. Xue et al. (2015) removed 24.7% of the NAs concentration in OSPW with an HRT of 48 days using a modified Ludzack-Ettinger membrane bioreactor (MLE-MBR) with a submerged ceramic membrane. Our previous study using moving bed biofilm reactors (MBBRs) showed a depletion of 34.8% of the NAs concentration in OSPW with an HRT of 48 days (Shi et al., 2015). The attached and/or suspended growth biomass employed in the above processes support a high bacteria retention time, encourage the growth of NAs removing microorganisms, and are more tolerant than planktonic bacteria to toxic substances (Davies, 2003; Sheng and Liu, 2011). Compared to activated sludge reactors, which are comprised of bacterial suspensions, a biofilm reactor can accumulate high concentrations of biomass by way of microorganism immobilization on carriers. Microorganisms with low specific growth rates can attach to the carriers and avoid being easily washed out (Gu et al., 2014). MBBRs are easy to operate and have been widely used in industrial wastewater treatment (Chen et al., 2007; Li et al., 2011).

In biological wastewater treatment, many microorganisms feed on the organic components in the water, thus removing pollutants. There is ample evidence that process performance is linked to the optimal microbial community structure in bioreactors (Konopka et al., 1999; Zhang et al., 2009). The operational conditions in a bioreactor should be carefully coordinated to control the growth of the microorganisms and to enhance their pollutant removal efficiency; the solids residence time (SRT), the HRT, nutrient loading rates, and nutrient ratios play significant roles in controlling an engineered bioreactor. The NAs in OSPW are toxic to humans and resistant to biodegradation. Therefore, factors that encourage the growth of microorganisms that remove NAs are the focus of much bioreactor engineering research. For example, a higher HRT can benefit the removal of resistant contaminants but demands an increased reactor volume. A lower HRT results in a higher organic loading rate, which leads to a higher biomass activity, but may also lead to incomplete organic biodegradation. Furthermore, the mechanisms of NAs biodegradation in OSPW are still unknown. As nitrifying bacteria have been shown to metabolize a variety of organic substances that typically resist biodegradation (Batt et al., 2006; Shi et al., 2011), nitrifying bacteria might play a key role during the NAs biodegradation. Therefore, we applied a strategy of decreasing C/N ratio by raising the influent ammonia concentration to enhance the nitrifying bacteria to see if this improved OSPW treatment.

Operational conditions of bioreactors affect biomass properties and microbial populations and thus affect the removal efficiency of contaminants from wastewater. Therefore in the present study we aimed to optimize the microbial populations to obtain optimal bioremediation, by changing the operational conditions (HRT and influent ammonia concentrations) in two MBBRs—one containing ozonated OSPW and one containing raw (no ozone treatment) OSPW. The toxicity of OSPW toward *Vibrio fischeri* was evaluated before and after MBBR treatment. Microbial gene abundance changes with parameter changes were measured for the evaluation of the overall performance. To investigate the relationships between bioreactor operational conditions (HRT and ammonia concentrations), microbial populations, and removal efficiency of OSPW helps us to obtain more information about the biodegradation of OSPW, which will be beneficial for developing the OSPW bioremediation strategies.

2. Material and methods

2.1. OSPW source and ozonation

Raw (no ozone pretreatment) OSPW and ozone treated OSPW were studied to assess the effect of ozonation on OSPW treatment. OSPW was collected from an oil sands tailings pond in Fort McMurray (Canada, in September 2013) and stored in polyvinyl chloride barrels before use (4 °C). Ozone-treated OSPW was obtained by ozonation (30 mg/L ozone dose) of OSPW using an ozone generator (WEDECO AG Water Technology, Herford, Germany), following the procedure of Shi et al. (2015). Characteristics of OSPW are listed in Table S1.

2.2. Reactor operation

Two MBBRs with a working volume of 8.5 L (15 cm × 35 cm base, 30 cm height), kindly provided by Napier-Reid Ltd. (Markham, Canada) were applied, one for raw OSPW treatment and one for ozonated OSPW treatment. Reactor parameters and operating conditions were similar to those in Shi et al. (2015). For reactors startup, a volume fraction of 60% carriers were introduced to the MBBR reactors. The reactors were operated continuously at room temperature (~23 °C). Air diffusers were applied for aeration and to continue the movement of carriers. DO was maintained between 6 and 8 mg/L. By the end of bioreactor startup, the biofilm thickness reached 97 ± 5 μm and 70 ± 12 μm for raw and ozonated OSPW systems respectively (Day 227). More data during the MBBRs startup could be found in the supporting information. In the present study, each reactor was optimized for OSPW treatment efficiency with respect to hydraulic retention time (HRT) and ammonium concentrations (Table 1). Extra carbon (200 mg/L COD), nitrogen, phosphorus, and other necessary nutrients were added to benefit the growth of bacteria in the MBBRs. Chemicals and supplies were obtained from Thermo Fisher Scientific.

2.3. Analyses of water quality

MBBR influent and effluent were collected and filtered (0.45 μm) before OSPW analysis. Chemical oxygen demand (COD) was tested using the standard method (Federation and American Public Health Association, 2005). Nitrate and ammonium concentrations were